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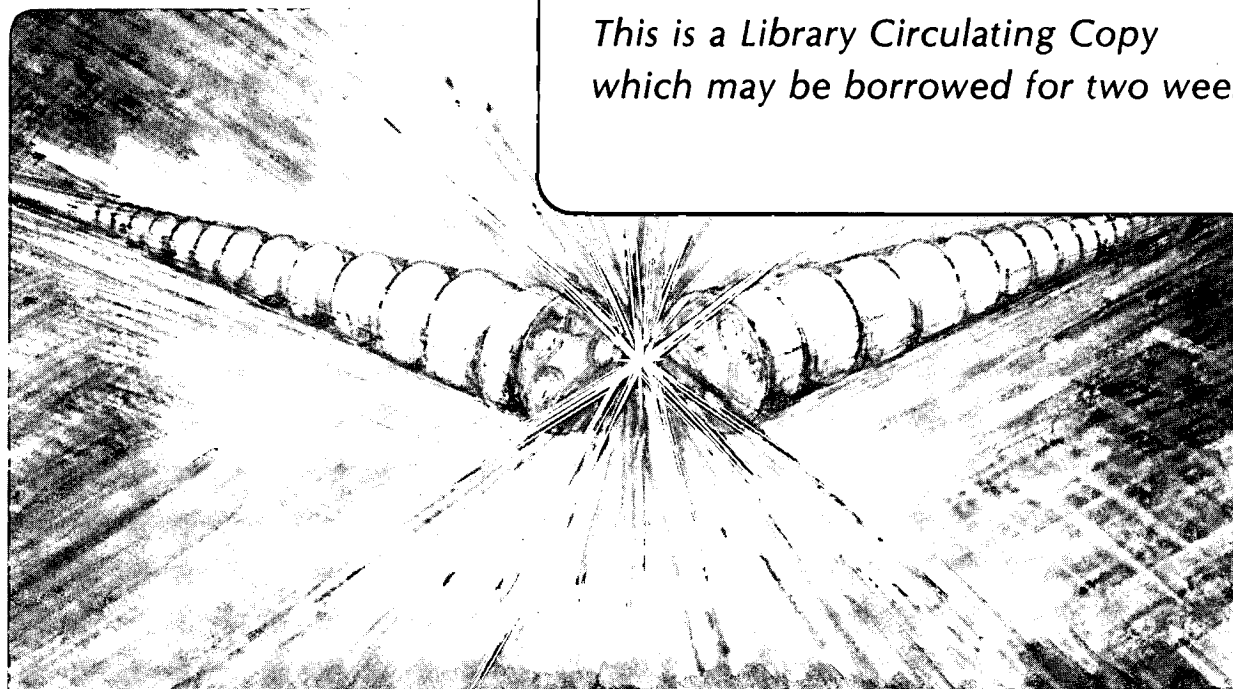
### The Kinetics of $H^-$ Ion Formation in Hydrogen Plasmas Studied Using Vacuum Ultraviolet Laser Absorption Spectroscopy

A.T. Young, G.C. Stutzin, A.S. Schlachter,  
K.N. Leung, and W.B. Kunkel

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THE KINETICS OF H<sup>-</sup> ION FORMATION IN HYDROGEN PLASMAS  
STUDIED USING VACUUM ULTRAVIOLET LASER ABSORPTION  
SPECTROSCOPY

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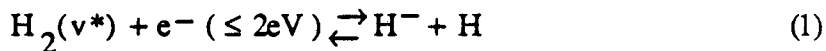
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# THE KINETICS OF H<sup>-</sup> ION FORMATION IN HYDROGEN PLASMAS STUDIED USING VACUUM ULTRAVIOLET LASER ABSORPTION SPECTROSCOPY

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The chemical kinetics occurring in medium electron-density hydrogen plasmas has been the subject of many recent investigations. In particular, the processes taking place in plasmas used as hydrogen ion sources have come under close scrutiny.<sup>1-2</sup> However, in spite of the recent work, the mechanism for negative-ion production is not well understood. One complicating factor in the study of the plasma chemistry is the difficulty of making measurements of the plasma constituents *in situ*, that is, in the plasma. This is due to the fact that the plasma produces a large photon and charged particle background which obscures the signal from most detection techniques. To overcome these difficulties and produce *in situ* measurements of H and H<sub>2</sub>, VUV laser absorption spectroscopy has been developed. This technique has been used to study the populations present in these plasmas under various conditions, revealing the kinetic processes taking place.

The most widely accepted model of H<sup>-</sup> formation in a "volume" source invokes the process of dissociative attachment.<sup>3</sup> In this process, vibrationally-excited molecules, H<sub>2</sub>(v\*), react with low-energy electrons to form H<sup>-</sup>, as shown in eqn. (1).



H<sub>2</sub> with  $v > 5$  are expected to be the most active species, as the cross sections for reaction (1) increase dramatically with vibrational level. In this scenario, the presence of highly-vibrationally-excited molecules is essential. The collision energy of the reactants is also important, with collisions of ~1 eV energy more reactive than collisions of  $\geq 10$  eV for producing H<sup>-</sup>. Finally, the reaction rate for the reverse reaction is also non-negligible and dependent on collision energy. Thus, determinations of the density and translational energy distribution of the H and H<sub>2</sub> are crucial for developing and testing the mechanisms of H<sup>-</sup> formation. VUV laser absorption has been used to measure the absolute, state-specific densities of these important species.

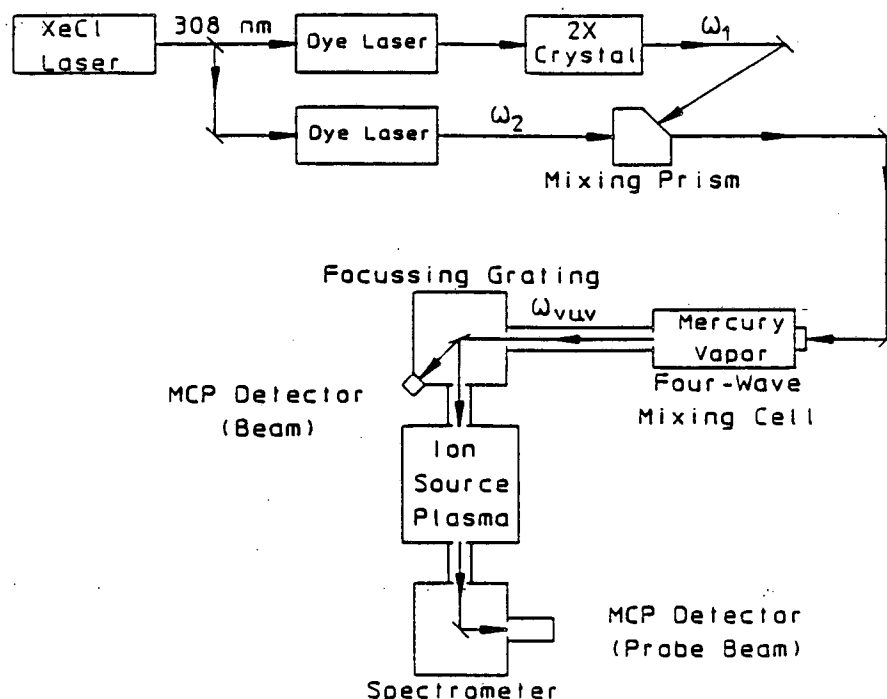
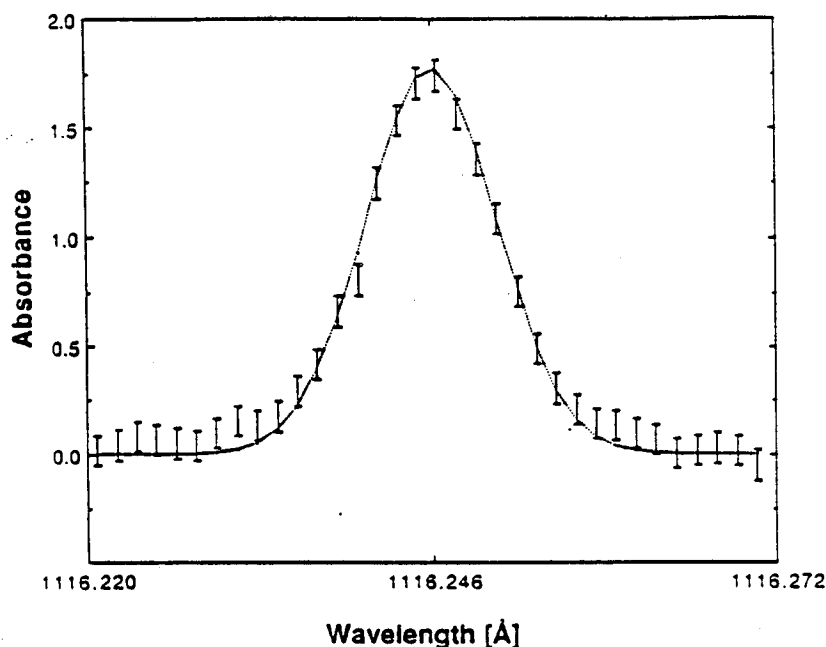


Figure 1. Experimental apparatus for vuv absorption spectroscopy.

The experimental apparatus has been previously described.<sup>4</sup> This is shown schematically in Fig. 1. Very briefly, narrow-band, directional, and continuously tunable VUV radiation is generated by focussing the high-intensity light from two excimer-pumped pulsed dye lasers into a mercury-vapor oven where four-wave sum frequency mixing takes place. The resulting VUV photon energy  $\epsilon_{vuv} = 2\epsilon_1 + \epsilon_2$  where  $\epsilon_1$  and  $\epsilon_2$  are the energies of the photons from the dye lasers. The VUV beam is split into two beams, one of which passes through the plasma of interest, and is measured with a microchannel-plate (MCP) detector. The other beam is measured without attenuation and is used for normalization. Using known values of the oscillator strength for  $H_2$  and H transitions allows us to compute the  $H_2$  and H concentrations in the plasma. The final  $H_2$  states for these measurements are specific rovibrational levels of either the  $B^1\Sigma_u^+$  or the  $C^1\Pi_u$  electronic states. For H detection, Lyman  $\beta(n=3 \leftarrow 1)$  or  $\gamma(n=4 \leftarrow 1)$  transitions are used.

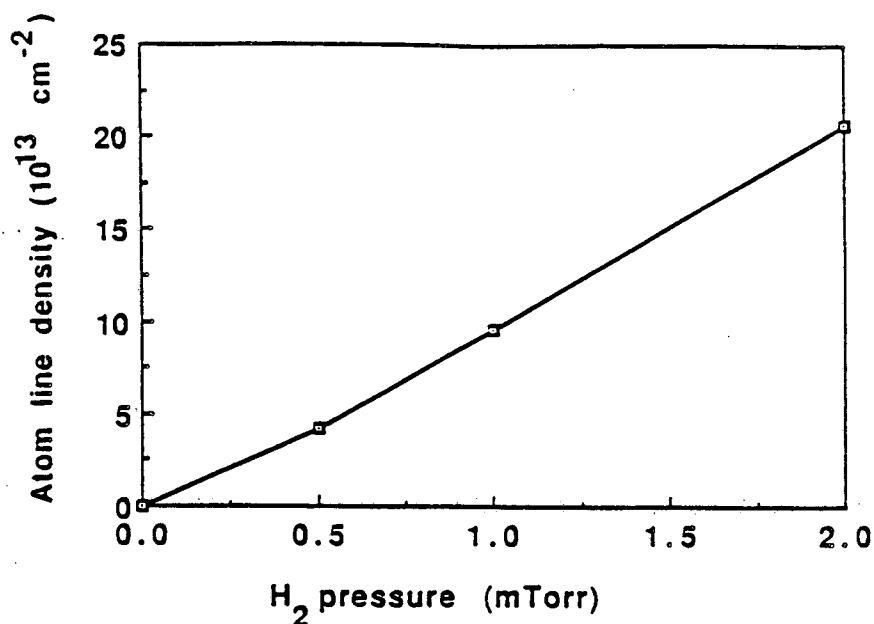


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Figure 2. VUV absorption spectrum of vibrationally-excited  $\text{H}_2$ . Transition observed is the (B-X) (3-1) R(2) line,

Figure 2 shows a typical absorption spectrum of  $\text{H}_2$ . This figure depicts the Lyman B  $1\Sigma_u^+ \leftarrow X\ 1\Sigma_u^+$  ( $v' = 3 \leftarrow v'' = 1$ ) R(2) transition. The absorption illustrated corresponds to a density of  $1.1 \times 10^{12}$   $\text{H}_2$  molecules  $\text{cm}^{-3}$  in this state. Also shown in Fig. 1 is a least-squares Gaussian fit to the data points. From the measured Doppler line width, we obtain a translation temperature of 0.037 eV (450 K). A detection limit of  $\sim 3 \times 10^9$  molecules- $\text{cm}^{-3}$ -quantum state has been achieved. Hydrogen atom densities of  $\sim 5 \times 10^{10}$  to  $1 \times 10^{14}$  have been measured with ion source discharge currents from 2 to 30 Amps and  $\text{H}_2$  pressures up to 10 mTorr. H-atom temperatures from 0.06 eV (700 K) to  $\sim 1$  eV (12000°K) have also been measured.

The H and  $\text{H}_2$  populations can be determined as functions of the plasma parameters. Figure 3 shows representative data for the variation of the measured H-atom line density with  $\text{H}_2$  pressure. The average H density is obtained by dividing the line density by the absorption path length, 30 cm. Data such as these are instrumental in determining the chemical processes important in these plasmas. In particular, vibrational distribution measurements of  $\text{H}_2$ , such as that in Figure 4, will provide key evidence for determining the key  $\text{H}^-$  formation reactions. These data represent the first measurements of  $\text{H}_2$  populations with  $v > 5$  in an ion source plasma, and has important implications for the proposed mechanism.



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Figure 3. Hydrogen atom line density in a plasma discharge as a function of initial H<sub>2</sub> pressure. Discharge conditions were 25A and 130V.

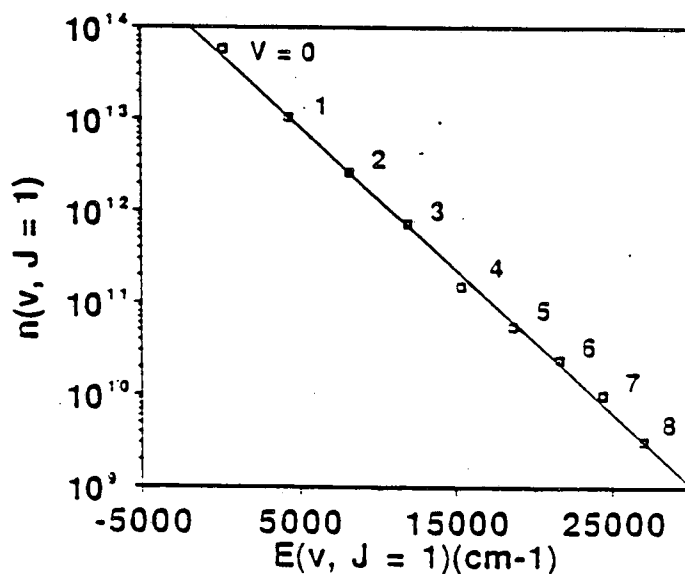


Figure 4. Vibrational state distribution of H<sub>2</sub> in ion source plasma.

In addition to complete H and H<sub>2</sub> measurements, the ion concentrations (e.g., H<sup>-</sup> and H<sup>+</sup>) will be determined mass spectrometrically. Comparison of the experimental data to the kinetic models of these plasmas will be made, and the implications to H<sup>-</sup> formation will be discussed.

## Acknowledgement

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